

Single shot measurement of light pulse duration by detection of excited acoustic wave

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Abstract. A new scheme for the measurement of picosecond and femtosecond single light pulses is proposed. The scheme is based on the generation of an acoustic wave with duration proportional to the light pulse but 10^3 – 10^4 times longer. Both volume and surface acoustic waves are considered.

1. Introduction

Considerable effort is now being made to obtain ultrashort laser pulses in the UV and VUV spectral ranges [1, 2]. One of the problems in this area is the measurement of the UV pulse duration, for which there are several reasons. First, the use of streak cameras is limited by the spectral sensitivity of the photocathode. Second, autocorrelation methods based on phase matched second harmonic generation [3, 4] also can not be used, because there are no crystals simultaneously able to be phase-matched below 400 nm and transparent below 200 nm. Other non-linear effects used to obtain a laser-pulse autocorrelation function are two-photon fluorescence [5] and multiphoton ionisation [6]. The acoustic detection of two-photon absorption of ultrashort pulses has been used to measure their duration in the 'many-shot' autocorrelation technique [7]. If the repetition rate of the ultrashort pulses is low and there are fluctuations in the pulse duration and energy, the best way to control the pulse duration is using a single-shot measurement technique. Here we both propose and consider theoretically single-shot duration measurements based on the excitation of an acoustic pulse by two-photon absorption.

2. Spatially inhomogeneous heating of matter by two-photon absorption process of colliding ultrashort pulses.

In order to simplify our analysis we consider only one of the possible mechanisms for sound generation by interband light absorption—thermoelastic excitation of acoustic waves due to inhomogeneous heating of the

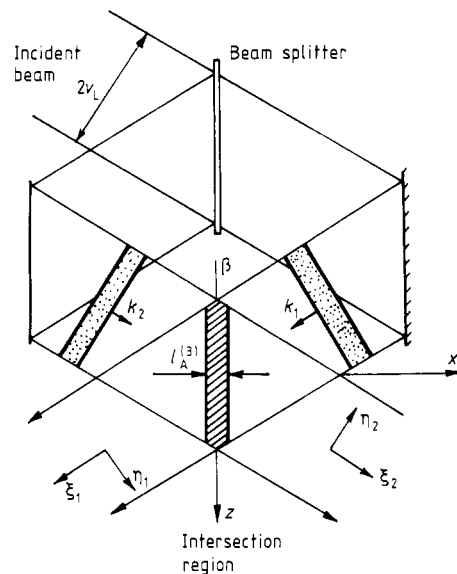


Figure 1. Schematic diagram of the interaction of two pulses in an acoustic medium. k_1 , k_2 are wavevectors, $l_A^{(3)}$ is characteristic length heated by the two-photon absorption.

sample [8]. Let us suppose that the light pulse is divided into two equal pulses (figure 1) with an angle 2β between their directions. We shall use the following expression for the electric field: $E_i = (0, E_i, 0)$

$$E_i = E_0 \exp(i\omega t - ik\xi_i) f\left(\frac{t - \xi_i/c_n}{\tau_L}\right) \varphi\left(\frac{\eta_i}{r_L}, \frac{y}{r_L}\right), \quad (1)$$

where we have separated the temporal dependence of the pulse (f) and the transversal distribution of the

field (φ). In equation (1)

$$\begin{aligned} \xi_i &= z \cos \beta \mp x \sin \beta \\ \eta_i &= x \cos \beta \pm z \sin \beta, \end{aligned}$$

r_L is the beam radius, $c_n = c/n$ is the light velocity, n is the index of refraction and τ_L is the light-pulse duration. The mutual disposition of the coordinate systems (x, y, z) , (ξ_1, y, η_1) and (ξ_2, y, η_2) is shown in figure 1. Using (1) one can obtain the square of the intensity distribution of the total electric field:

$$I^2 \sim [(E_1 + E_2)(E_1 + E_2)^*]^2$$

i.e.,

$$\begin{aligned} I^2 &= I_0^2 \{ (f_1 \varphi_1)^4 + (f_2 \varphi_2)^4 + 4(f_1 \varphi_1 f_2 \varphi_2)^2 \\ &+ 2(f_1 \varphi_1 f_2 \varphi_2)^2 \cos 4\omega\gamma x + 4[f_1 \varphi_1 (f_2 \varphi_2)^3 \\ &+ (f_1 \varphi_1)^3 f_2 \varphi_2] \cos 2\omega\gamma x \}. \end{aligned} \quad (2)$$

Here, $\gamma = c_n^{-1} \sin \beta$ and I_0 is the peak intensity of the pulse.

The medium is heated due to both linear and two-photon absorption processes. Because of the non-linear intensity dependence of the absorption, the intersection region absorbs more light energy. Some reasonable assumptions can now be made. If the characteristic cooling time of the intersection region τ_T is long in comparison with the duration of the excited acoustic pulse τ_A ($\tau_T \gg \tau_A$), then thermal conductivity in the evaluation process of the temperature field can be neglected [8, 9, 10]. We also suppose that the thermalisation time of the absorbed light energy τ_R is much shorter than the acoustic pulse duration ($\tau_R \ll \tau_A$), and this means that the process of lattice heating can be considered as instantaneous [9, 10]. Thus, neglecting the delay in heating due to the finite time of electron-hole pair recombination and omitting radiative recombination [10], one can describe the temperature rise caused by two-photon interband light absorption in a rather simple way:

$$T(x, y, z) = \frac{k_2}{\rho_0 c_p} \int_{-\infty}^t I^2(x, y, z, t') dt'. \quad (3)$$

Here ρ_0 is the density of the crystal; c_p the specific heat and k_2 the two-photon absorption coefficient. We have deliberately omitted the linear absorption contribution to the temperature field, supposing that I_0 is so high that

$$k_2 I_0 \equiv \alpha_{NL} \gg \alpha. \quad (4)$$

If we take into account that

$$\tau_A \gg \tau_L \quad (5)$$

the net heating of the medium will be

$$T = \frac{k_2}{\rho_0 c_p} \theta(t) \int_{-\infty}^{+\infty} I^2(x, y, z, t') dt' \quad (6)$$

where

$$\theta(t) = \begin{cases} 1 & t \geq 0 \\ 0 & t < 0. \end{cases}$$

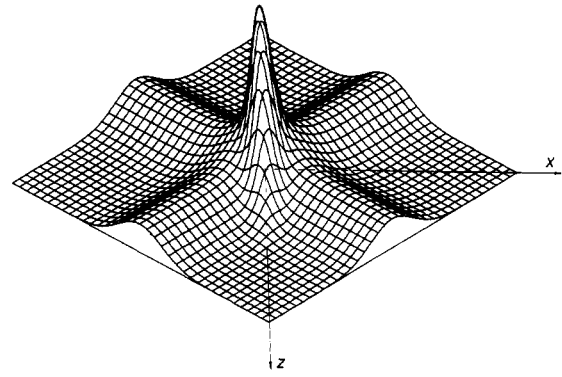


Figure 2. Calculated temperature field in the acoustic medium. A Gaussian shape for the temporal and spatial profile of the pulses is assumed.

Substituting (2) into (6) one obtains the result

$$\begin{aligned} T &= T_0 \theta(t) \left[(\varphi_1^4 + \varphi_2^4) \int_{-\infty}^{+\infty} d\sigma f^4(\sigma) \right. \\ &+ 4 \cos 2\omega\gamma x (\varphi_1^3 \varphi_2 + \varphi_1 \varphi_2^3) \int_{-\infty}^{+\infty} d\sigma f^3 \left(\sigma + \frac{x\gamma}{\tau_L} \right) f \\ &\times \left(\sigma - \frac{x\gamma}{\tau_L} \right) + 2(2 + \cos 4\omega\gamma x) (\varphi_1 \varphi_2)^2 \\ &\left. \times \int_{-\infty}^{+\infty} d\sigma f^2 \left(\sigma + \frac{x\gamma}{\tau_L} \right) f^2 \left(\sigma - \frac{x\gamma}{\tau_L} \right) \right] \quad (7) \end{aligned}$$

where $T_0 = k_2 I_0 \tau_L / \rho_0 c_p$.

The calculated temperature field T for a Gaussian shape for φ and f is presented in figure 2. Inhomogeneous heating of the medium given by (7) leads to sound generation. Two heated tubes along the propagation directions of the light pulses (terms proportional to φ_1^4 and φ_2^4 in (7)) excite cylindrical acoustic waves with characteristic spatial length $l_A^{(1)} (l_A^{(1)} \sim r_L)$. In the intersection area two other acoustic waves are excited. One of them is caused by an interference pattern and has a characteristic length $l_A^{(2)} \sim 1/\omega\gamma$. In this paper we shall not consider this high-frequency acoustic wave. We suppose that it will be absorbed before reaching the detector or that the detector does not have sufficient resolution to detect it. The heating intersection region will also generate an acoustic pulse with a characteristic length $l_A^{(3)} \sim (\tau_L/\gamma) \gg l_A^{(2)}$. Registration of the shape of this sound pulse could be used for reconstruction of the light pulse shape and duration.

In the next two sections we consider two possible experimental schemes for registration of the acoustic pulse with characteristic spatial length $l_A^{(3)}$. In choosing these schemes two factors have been kept in mind—elimination of parasitic acoustic signals on the acoustic detector and to minimisation of additional processing of the acoustic signal for the reconstruction of the temporal shape of the light-pulse. The first scheme is based on the longitudinal acoustic (LA) volume pulse generation and the other on Rayleigh surface acoustic wave (SAW) excitation.

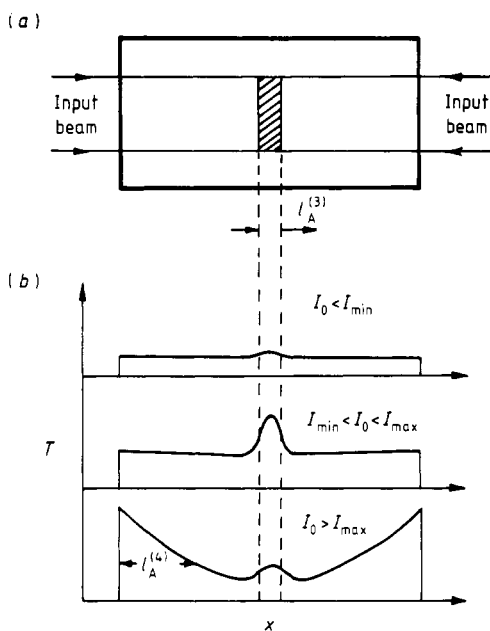


Figure 3. The influence of intensity of the interacting pulses on the temperature field in the scheme when two pulses are propagating in opposite directions.

3. Measurement of light-pulse duration by LA pulse registration

Let us consider two light pulses propagating in opposite directions as shown in figure 3(a) ($\beta = \pi/2$, $\varphi_1 = \varphi_2 = \varphi(r/r_L)$, $r = (z^2 + y^2)^{1/2}$). Then (7) can be transformed to

$$T = 2T_0\varphi^4\theta(t) \left[\int_{-\infty}^{+\infty} d\sigma f^4(\sigma) + 2 \int_{-\infty}^{+\infty} d\sigma f^2 \times \left(\sigma + \frac{x}{c_n\tau_L} \right) f^2 \left(\sigma - \frac{x}{c_n\tau_L} \right) \right] \equiv (T'(r) + T''(r, x))\theta(t). \quad (8)$$

The term T'' is proportional to the second-order time autocorrelation function $G^{(2)}$ of the light pulse. The width of $G^{(2)}$ is $S_G = l_A^{(3)} = c_n\tau_L/2\kappa$, where $\kappa = \tau_L/\tau_G$, is the form factor given in [11] for ten different pulse shapes.

In the case of $l_A^{(1)} \gg l_A^{(3)}$ the inhomogeneous temperature field $T''(r, x)\theta(t)$ is a source of quasiplanar acoustic waves. The generation and propagation of these acoustic waves up to distances x shorter than the diffraction length l_D ($l_D \sim (l_A^{(1)})^2/l_A^{(3)}$) may be described by the one-dimensional wave equation:

$$U_{tt} - c_{LA}^2 U_{xx} = -\frac{B\beta}{\rho_0} T_x U|_{t=0} = 0 \quad (9)$$

where U is the displacement of the particles, c_{LA} is the speed of the longitudinal acoustic waves, B is the bulk elastic modulus and β is the volume coefficient of thermal expansion. The exact solution of equation (9) gives two longitudinal deformation U_x pulses escaping from

the heated area in opposite directions:

$$U_x = \frac{B\beta}{\rho_0 c_{LA}^2} \frac{T''(r, x - c_{LA}t) + T''(r, x + c_{LA}t)}{2} \theta(t). \quad (10)$$

The profiles of the generated acoustic pulses (10) follow the temperature distribution T'' in (8). Thus their duration is equal to the width of the autocorrelation function and from this width, assuming some shape for the light pulse, its duration could be extracted [11]. In fact we expand the measured ultrashort pulse, transforming it into the nanosecond scale. The coefficient of expansion

$$M_{LA} = \frac{\tau_{LA}}{\tau_L} = \frac{c}{\kappa n c_{LA}} \quad (11)$$

depends on the pulse shape. For a Gaussian pulse shape $\kappa = 1/\sqrt{2}$. Since $c_n \gg c_{LA}$ then $\tau_{LA} \gg \tau_L$ and therefore assumption (5) is confirmed. In solids the coefficient $M_{LA} \sim 10^4$, but in liquids and gases it takes even higher values.

Characteristic pressures p and displacements U in the acoustic pulse can be estimated as follows:

$$|p| \sim \rho_0 c_{LA}^2 |U_x| \sim B\beta T_0 \quad (12)$$

$$|U| \sim |U_x| c_{LA} \tau_{LA} \sim \frac{|p|}{\rho_0 c_{LA}} \tau_{LA}.$$

when making estimates one should not forget assumption (4), which imposes a lower limit on the possible light intensities $I_0 \geq \alpha/k_2 = I_{\min}$. On the other hand, if the light intensity is too high, two-photon absorption of each pulse will reduce the intensity of the pulses before their meeting and non-uniform heating of the medium will occur along the path of the pulse with characteristic length $l_A^{(4)} \sim \alpha_{NL}^{-1}$. As a result, an acoustic pulse with duration $l_A^{(4)}/c_{LA}$ is excited. To avoid such an effect the intensity must be kept below I_{\max} so that $l_A^{(4)} > l_A^{(3)}$ or $I_0 \leq (3k_2 c_{LA} \tau_{LA})^{-1} \equiv I_{\max}$. An illustration of the temperature field for different I_0 is shown in figure 3. It is seen that for $I_0 > I_{\max}$ the amplitude of the informative acoustic pulse, i.e. the one with $l_A^{(3)}$, is reduced.

In table 1 the results of our numerical estimates are presented, together with the used values of the physical constants for GaAs, NaCl, H₂O and SiO₂. The estimated pressures and displacements are quite detectable. For comparison we note that the piezoelectric transducers based on lead zirconate titanate have a sensitivity of 3V bar⁻¹ [16]. Other data on sensitive techniques for acoustic pulse registration can be found in [17–20].

Two possible schemes for experimental realisation of the proposed acoustic method are shown in figure 4.

4. Measurement of light pulse duration by Rayleigh saw registration

Let us investigate SAW generation in the following experimental conditions: $z < 0$ for vacuum (or air),

Table 1. Physical quantities (some from the references indicated; others are well known physical constants) and numerical estimates from our theoretical predictions for GaAs, NaCl, H₂O and SiO₂ (denoted by †).

Parameter	GaAs	NaCl	H ₂ O	SiO ₂								
λ (μm)	1.06	0.266	0.266	0.266								
n	3.5	1.66	1.4	1.5								
α (cm^{-1})	0.55 [12]	0.23 [13]	0.013	$\sim 2 \times 10^{-3}$								
K_2 (cm GW^{-1})	26 [12]	9.2 [13]	0.1 [14]	0.045 [15]								
ρ_0 (g cm^{-3})	5.3	2.2	1	2.65								
c_p ($\text{J g}^{-1} \text{K}^{-1}$)	0.35	0.85	4.2	0.74								
$\langle \text{direction} \rangle$	$\langle 111 \rangle$	$\langle 100 \rangle$		$\langle 010 \rangle$								
c_{LA} (10^5 cm s^{-1})	5.4	4.7	1.5	5.4								
$\langle \text{direction} \rangle$	$\langle 111 \rangle$	$\langle 100 \rangle$		$\langle 010 \rangle$								
c_{TA} (10^5 cm s^{-1})	2.8	2.4	—	3.8								
$B = \rho_0(c_{\text{LA}}^2 - \frac{1}{3}c_{\text{TA}}^2)$ (10^5 bar)	9.9	3.2	0.225	2.5								
β (10^{-5} K^{-1})	1.6	12	30	5.0								
† $M_{\text{LA}} = \frac{\sqrt{2}c}{n c_{\text{LA}}}$	2.2×10^4	5.6×10^4	1.9×10^5	4.7×10^4								
† I_{min} (MW cm^{-2})	21	25	130	44								
τ_{L} (ps)	10	1	0.1	10	1	0.1						
† τ_{LA} (ns)	220	22	2.2	560	56	5.6	1900	190	19	480	48	4.8
† I_{max} (GW cm^{-2})	0.3	3	30	1	10	100	30	300	3000	80	800	8000
† $ \rho $ ($I = I_{\text{max}}$) (bar)	0.2	2	20	0.7	7	70	1.5	15	150	2	20	200
† $ U $ ($I = I_{\text{max}}$) (Å)	0.6	0.6	0.6	14	14	14	700	700	700	23	23	23

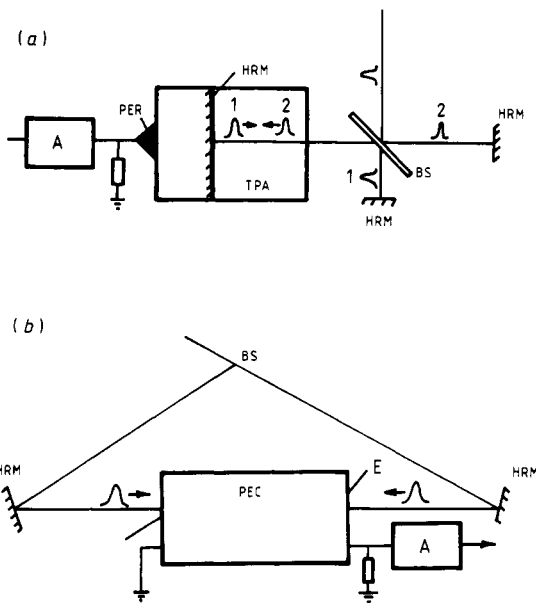


Figure 4. Two possible schemes for experimental realisation of light-pulse duration measurement by longitudinal acoustic pulse registration: (a) one-side scheme and (b) two-side scheme with a piezoelectric crystal (PEC) as a two-photon absorption medium. BS: beamsplitter, PER: pyroelectric receiver, TPA: two-photon absorption medium, HRM: high-reflection mirror, A: amplifier, E: electrodes.

$0 < z < a$ for a crystal film with high two-photon light absorption, $z > a$ for an optically transparent substrate acoustically matched with the film. Using (1) the light

intensity distribution on the film surface can be represented in the form

$$I(z=0) = I_0 \varphi^2 \left(\frac{x \cos \beta}{r_L}, \frac{y}{r_L} \right) \left[f^2 \left(\frac{t + \gamma x}{\tau_L} \right) + f^2 \left(\frac{t - \gamma x}{\tau_L} \right) + 2f \left(\frac{t + \gamma x}{\tau_L} \right) f \left(\frac{t - \gamma x}{\tau_L} \right) \cos(2\omega \gamma x) \right]. \quad (13)$$

Some characteristic lengths of the intensity modulation along the x axis can be introduced: $l_{\text{RA}}^{(1)} \sim r_L / \cos \beta$, $l_{\text{RA}}^{(2)} \sim c / \omega \sin \beta$, $l_{\text{RA}}^{(3)} \sim \tau_L c / \sin \beta$. We again neglect high-frequency modulation (as was done in § 3).

Let us now assume that

$$\min(\alpha_{\text{NL}}^{-1}, a) \ll \min(l_{\text{RA}}^{(1)}, l_{\text{RA}}^{(3)}). \quad (14)$$

Then the intensity distribution in the film is described by the one-dimensional equation

$$I_z = -k_2 I^2.$$

Its exact solution is

$$I(x, y, z, t) = \frac{I(z=0)}{1 + k_2 I(z=0)z}. \quad (15)$$

Substitution of (15) into (6) gives the spatially inhomogeneous temperature rise in the film.

If the light beam radius is so large that $r_L \gg l_{\text{RA}}^{(3)}$, weakly diffracting SAWs will be excited and propagated along x in opposite directions. According to [9], for

the perpendicular to the surface displacement U_{\perp}

$$U_{\perp}\left(\theta_s = t - \frac{x}{c_{RA}}\right) = \frac{A}{2\pi} \int_{-\infty}^{+\infty} d\Omega \exp(-i\theta_s \Omega) \hat{T}_t\left(\frac{\Omega}{c_{RA}}, y, \frac{|\Omega|}{c_{RA}} a_2, \Omega\right) \quad (16)$$

is valid, where

$$\begin{aligned} \hat{T}_t(q, y, p, \Omega) &= \iint_{-\infty}^{+\infty} dx dt \int_0^{+\infty} dz \\ &\times \exp(i\Omega t - iqx - pz) T_t(x, y, z, t), \\ A &= -\frac{B\beta c_{RA}}{16\rho_0 c_{LA}^2 c_{TA}^2} \left(\frac{a_1^2 + a_2^2 + 2a_1^2 a_2^2}{4(a_1 a_2)^{3/2}} - 1\right)^{-1} \\ a_1 &= [1 - (c_{RA}/c_{TA})^2]^{1/2} \\ a_2 &= [1 - (c_{RA}/c_{LA})^2]^{1/2}. \end{aligned}$$

In the above expressions c_{RA} is the SAW velocity and c_{TA} is the volume velocity for shear waves.

Using (3) and (15) and performing a Laplace transformation in (16) we arrive at

$$\begin{aligned} \int_0^{+\infty} dz T_t(x, y, z, t) \exp(-pz) &= F \\ &= \frac{k_2}{\rho_0 c_p} \int_0^a I^2(x, y, z, t) \exp(-pz) dz \\ &= \frac{I(z=0)}{\rho_0 c_p} \left(1 - \frac{\exp(-pa)}{1+b} - \frac{pa}{b} \int_0^b \frac{\exp(-pa\xi/b)}{1+\xi} d\xi\right) \end{aligned} \quad (16a)$$

where $b = ak_2 I(z=0)$. From formula (16) it is obvious that the wavevector of the SAW is $p \sim |\Omega|/c_{RA}$. In the considered case $p \leq 1/\min(I_{RA}^{(1)}, I_{RA}^{(3)})$ so it can be easily shown using (14) and (16a) that $b \gg 1$ ($a \gg \alpha_{NL}^{-1} \sim (k_2 I_0)^{-1}$), i.e. for a 'thick' film we have

$F \approx I(z=0)/\rho_0 c_p$. In other words, the profile of the SAW is related to the intensity distribution on the surface which carries information about the first-order autocorrelation function $G^{(1)}$ of the light pulse. This autocorrelation could not be used for correct determination of the light-pulse duration. To extract information for τ_L from low-frequency SAW one has to satisfy experimentally the condition $a \ll \alpha_{NL}^{-1}$ in order to use a 'thin' film. Then

$$F \approx \frac{ak_2 I^2(z=0)}{\rho_0 c_p}$$

and for the displacement of the surface U_{\perp} we obtain formula similar to (8):

$$\begin{aligned} U_{\perp} &= 2AT_0 c_{RA} a \varphi^4 \left(\frac{\theta_s c_{RA} x \cos \beta}{r_L}, \frac{y}{r_L}\right) \\ &\times \left[\int_{-\infty}^{+\infty} d\sigma f^4(\sigma) + 2 \int_{-\infty}^{+\infty} d\sigma f^2 \left(\sigma - \frac{\theta_s \gamma c_{RA}}{\tau_L}\right) \right. \\ &\left. \times f^2 \left(\sigma + \frac{\theta_s \gamma c_{RA}}{\tau_L}\right) \right]. \end{aligned} \quad (17)$$

Two acoustic pulses with different characteristic lengths are excited. In order to distinguish them we should choose the ratio of their durations to be at least 3, which is equivalent to the following restriction for the beam radius at fixed angle β between the beams:

$$r_L \geq (3\tau_L c)/\sin \beta. \quad (18)$$

The coefficient of expansion M_{RA} increases with decreasing β :

$$M_{RA} = c/\kappa c_{RA} \sin \beta. \quad (19)$$

We could not reduce β infinitely because in order to fulfill (18), r_L should be increased and this will lead to a decrease in the light-pulse intensity and consequently in the amplitude of the informative acoustic wave. Another advantage of this scheme in comparison

Table 2. Estimation of the SAW characteristics (\dagger) in GaAs and NaCl for three different values of the light pulse durations. For each of them an appropriate value for the pulse energy is chosen.

Parameter	GaAs			NaCl		
λ (μm)	1.06			0.266		
$c_{RA} \approx 0.9c_{TA}$ (10^5 cm s^{-1})	2.5			2.2		
$\dagger M_{RA}^{(\text{min})} \times \sqrt{2}c/c_{RA}$	17 10^4			19 10^4		
τ_L (ps)	10	1	0.1	10	1	0.1
M_{RA}		20 $\times 10^4$			20 $\times 10^4$	
$\dagger \tau_A$ (ns)	2000	200	20	2000	200	20
$\dagger \beta = \sin^{-1} \left(\frac{M_{\text{min}}}{M}\right)_{RA}$ (deg)	58.2			71.8		
$\dagger r_L^{\text{min}} = 3\tau_L c/\sin \beta$ (cm)	1.1	0.11	0.011	0.95	0.09	0.009
W_0 (energy in light pulse) (mJ)	10	0.5	0.01	10	0.5	0.01
$\dagger I_0 \sim \frac{W_0}{\tau_L r_L^2}$ (GW cm^{-2})	0.8	41	826	1.2	60	1200
$\dagger a \approx 0.1 \alpha_{NL}^{-1}$ (μm)	48	0.9	0.5	90	1.8	0.9
$\dagger U_{\perp} $ (\AA)	0.25	1.3	2.6	3	14	28

with the scheme using longitudinal acoustic waves, is the absence of an upper limit for the intensities I . We may increase I_0 and simultaneously decrease the thickness of the film (fulfilling, for example, $a \approx 0.1\alpha_{NL}^{-1}$) and then the acoustic signal will increase proportionally with I_0 .

In table 2 estimates of SAW characteristics in GaAs and NaCl films for three different pulse durations are listed. The estimated acoustic signal can be registered by optical methods [21] or by piezoelectric transducers [22].

5. Conclusion

We have shown the potential for single-shot measurement of picosecond and femtosecond light pulses using acoustic-wave excitation. From our estimate it is seen that a light pulse of 100 fs duration and 10 μ J energy is easily detectable. This is several orders of magnitude lower than the energy values quoted in [1, 23].

In the present paper, high-frequency modulation of the acoustic pulses has been neglected and hence only intensity autocorrelation functions can be reconstructed. If one takes into account the interference pattern and can register it, more information about the light pulse, e.g. the chirp, could be extracted from the phase autocorrelation function [24].

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